

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of:

Christoph Briehn et al.

Group Art Unit: 4171

Examiner: Hannah J. Pak

Serial No.: 10/599,285

Filed: September 25, 2006

For: CURABLE COMPOSITION CONTAINING  
SURFACE-MODIFIED PARTICLES

Attorney Docket No.: WAS 0807 PUSA

**DECLARATION OF DR. CHRISTOPH BRIEHN  
UNDER RULE 37 C.F.R. 1.132**

Commissioner for Patents  
United States Patent and Trademark Office  
Washington, D.C. 20231

Sir:

I, Dr. Christoph Briehn, do hereby declare and state as follows:

1. I am a co-inventor of the subject matter disclosed and claimed in U.S. Application Serial No. 10/599,285, and am familiar with the contents of the application, including the claims, the Office Actions received, and the cited prior art, *Olsen et al.* U.S. Patent 4,491,508, which was discussed on page 1 of the subject application.

2. The scratch resistance of coatings has been a long recognized problem in the coatings industry. *Olsen* teaches the addition of silica modified with (meth)acrylatoalkylenealkoxysilanes where the alkylene group has 4 to 10 carbon atoms ( $R^3$  is  $C_{1-8}$  alkyl). The *Olsen* patent issued in 1985, and despite the knowledge of its issuance more than 20 years ago, the coatings art has still sought for coatings with improved scratch resistance.

3. Applicants solved this long felt need by functionalizing silica with (meth)acrylatomethylalkoxysilanes. As a result of the use of these compounds, where the silicon of the alkoxysilyl group is separated from the oxygen of the (meth)acrylato group by but a single methylene group, the scratch resistance was very surprisingly and unexpectedly increased by a significant amount.

4. To further illustrate the surprising and unexpected nature of the claimed invention, further examples were performed under my direction in the Assignee's laboratories in Burghausen, Germany. A further inventive example, using the claimed methacrylatomethyltrimethoxysilane was compared with two comparative examples using methacrylatopropyltrimethoxysilane, one example using ethanol as the solvent (Comp. Ex. B), the same as in the inventive example, so as to be directly comparable, and one employing the solvent used by *Olsen*, t-butanol. The experimental details are as follows, and the scratch resistance results are presented in the Table:

New Inventive Example:

20.00 g of an aqueous SiO<sub>2</sub> sol (LUDOX® AS 40 from Grace Davison, 40% by weight SiO<sub>2</sub>, pH = 9.1, 22 nm) are admixed dropwise over the course of 60 minutes with 20 ml of ethanol and over 5 minutes with 2.00 g of methacrylatomethyltrimethoxysilane and the mixture is heated at 60°C for 16 hours. After the mixture is cooled to room temperature, 15.00 g of hexanediol diacrylate are added and then ethanol and water are distilled off as an azeotrope. The transparent dispersion contains 35% by weight of SiO<sub>2</sub>.

New Comparative Example A:

20.00 g of an aqueous SiO<sub>2</sub> sol (LUDOX® AS 40 from Grace Davison, 40% by weight SiO<sub>2</sub>, pH = 9.1, 22 nm) are admixed dropwise over the course of 60 minutes with 20 ml of tert.-butanol and over 5 minutes with 2.00 g of methacrylatopropyltrimethoxysilane and the mixture is heated at 60°C for 16 hours. After the mixture is cooled to room temperature, 15.00 g of hexanediol diacrylate are added and then tert.-butanol and water are distilled off as an azeotrope. The transparent dispersion contains 35% by weight of SiO<sub>2</sub>.

## New Comparative Example B:

20.00 g of an aqueous SiO<sub>2</sub> sol (LUDOX® AS 40 from Grace Davison, 40% by weight SiO<sub>2</sub>, pH = 9.1, 22 nm) are admixed dropwise over the course of 60 minutes with 20 ml of ethanol and over 5 minutes with 2.00 g of methacrylatopropyltrimethoxysilane and the mixture is heated at 60°C for 16 hours. After the mixture is cooled to room temperature, 15.00 g of hexanediol diacrylate are added and then ethanol and water are distilled off as an azeotrope. The transparent dispersion contains 35% by weight of SiO<sub>2</sub>.

Coatings were prepared and evaluated according to Example 6 and Example 7 of the patent application.

The scratch resistance of the new inventive example and the new comparative examples is as follows:


| Coating Sample             | Loss of Gloss |
|----------------------------|---------------|
| New Inventive Example      | 19%           |
| New Comparative Example A  | 23%           |
| New Comparative Example B  | 24%           |
| 1, 6-Hexanediol Diacrylate | 73%           |

Table 1: Loss of gloss in the Peter-Dahn scratch test

5. Compared with the subject inventive example with a loss of gloss of only 19%, the two comparative examples had loss of gloss of 23% and 24%, respectively. On a relative basis, therefore, the subject invention coating had a scratch resistance higher by 21% and 26%, respectively. This increase in scratch resistance is highly surprising and unexpected, and could not be predicted. This increase is also very commercially significant, where even small increases in scratch resistance are considered of high importance. In the more than 20 years since the *Olsen* patent was published, no one has sought to use the functionalizing silanes used by Applicants, despite the long felt need for higher scratch resistance.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

  
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Dr. Christoph Brähn

Dated: 29.05.2009